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Spatially resolved optical and electro-optic properties
of electroclinic liquid crystals

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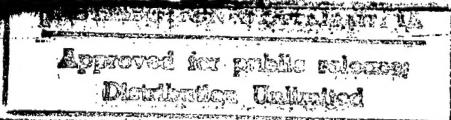
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ABSTRACT

Chiral smectic A (Sm A) liquid crystals are of increasing interest because of their potential for fast analog electrooptic modulation. Chiral smectic A liquid crystals, when viewed between crossed polarizers, exhibit distinctive periodic stripe domains due to a voltage dependent deformation of the bookshelf geometry. These stripe domains reduce the contrast ratio of electrooptic devices employing these materials. This paper describes a novel optical technique to probe spatial variations in the optical properties of liquid crystals with micron resolution. The effect of the stripe formation on the electrooptic properties of a electroclinic Sm A liquid crystal, denoted as KN125, is investigated.

The electric field induced stripe domains were experimentally characterized by measuring the transmission of KN125, interposed between crossed polarizers. A sharply focussed visible cw laser beam was translated across the sample with one-micron resolution. A quasi periodic modulation of the transmission due to stripe deformation of the bookshelf geometry was observed. Measurements performed within a single stripe and between adjacent stripes permitted an independent determination of the true optical tilt angle and that of the stripe deformation. The electric field dependence of the optical tilt angle for the KN125 liquid crystal is reported.

The electroclinic effect in chiral smectic A liquid crystals has attracted considerable recent attention because of the large field-induced molecular tilt angles possible in these materials and their potential use in applications requiring a fast linear electrooptic response and analog phase modulation. One application for chiral liquid crystals in their smectic A phase is that of high speed two dimensional spatial light modulators with grey scale operation. However, when chiral smectic A liquid crystals with a large electroclinic effect are subjected to an electric field and viewed between crossed polarizers, they exhibit distinctive periodic stripe domains due to a voltage deformation of the bookshelf geometry. The resulting molecular alignment alternates between stripes, reducing the contrast ratio and imposing performance limitations on electrooptic devices employing these materials. A greater understanding of the nature and origin of the stripe domains is needed, for both fundamental and practical concerns.



X-ray diffraction studies reveal that the distribution of the layer normals of the stripes are nearly bimodal indicating that the layer distortions possess a triangular profile¹ as predicted by Pavel and Glogarva.² In the present investigation, the stripe deformation is spatially resolved using a novel technique to probe the electrooptical properties with micron resolution. Resolution of the spatial variation in the molecular alignment allows a more thorough evaluation of the overall performance of the material as well as measurement of its electroclinic properties.

This study was performed on the chiral smectic A liquid crystal KN125, which was synthesized at NRL and shown to exhibit a fast electroclinic behavior. The structure of the KN125 molecule is shown in figure 1. The phase transition sequence³ for KN125 is as follows: crystal - (33 °C) - chiral smectic A - (78 °C) - isotropic. KN125 is transparent in the visible spectral region.

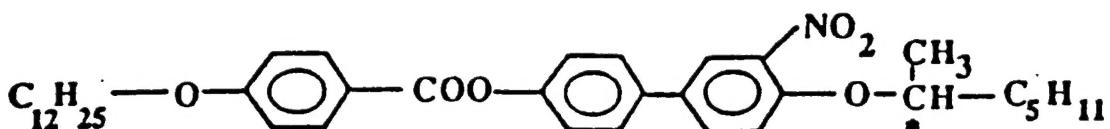


Figure 1. The molecular structure of the Liquid Crystal, KN125.

This compound was loaded into prefabricated glass cells whose thickness had been determined interferometrically. The interior surfaces of the cells were coated with indium tin oxide (ITO) transparent electrodes, and with a polyimide overlayer which was mechanically rubbed to facilitate uniform alignment. The compound was heated above the isotropic phase (~85 °C) allowing the cells to fill by capillary action. The temperature was lowered below the isotropic phase transition and a small electric field was applied to align the liquid crystal. The electric field was removed and the compound was slowly cooled to room temperature where KN125 supercools in the chiral smectic A phase and remains stable for months. Prior to performing the spatially resolved measurements, the sample was subjected to electric fields up to 10 V/ μ m and the existence of stripe domains was verified by optical microscopy.

The electric-field-induced stripe domains were experimentally characterized using the apparatus diagrammed in Figure 2. The output of either a tunable Ar^+ ion or He-Ne laser was expanded to fill the aperture of the Glan polarizer and directed through a half wave plate. (A fixed-wavelength half-wave plate or a Babinet-Soliel compensator set to half wave retardation at the laser wavelength was employed.) The half wave retarder was mounted on a computer controlled, high resolution ($\pm 0.001^\circ$) rotation stage as was the analyzing Glan polarizer behind the sample. The waveplate and analyzer were rotated in tandem in order to maintain precise beam and sample alignment. The liquid crystal cell was mounted on a high precision (1 μ m) XYZ translation stage with the transverse, X, direction under computer control.

The optical beam was focussed onto the KN125 sample using high quality f/5 optics. The sample was precisely positioned in the focal plane, and the axis of translation carefully oriented perpendicular to the optical beam to maintain the sample at focus. Focal spot diameters of 4 μ m and 5 μ m at 488.0 nm and 632.8 nm, respectively, were determined by a knife-edge technique.

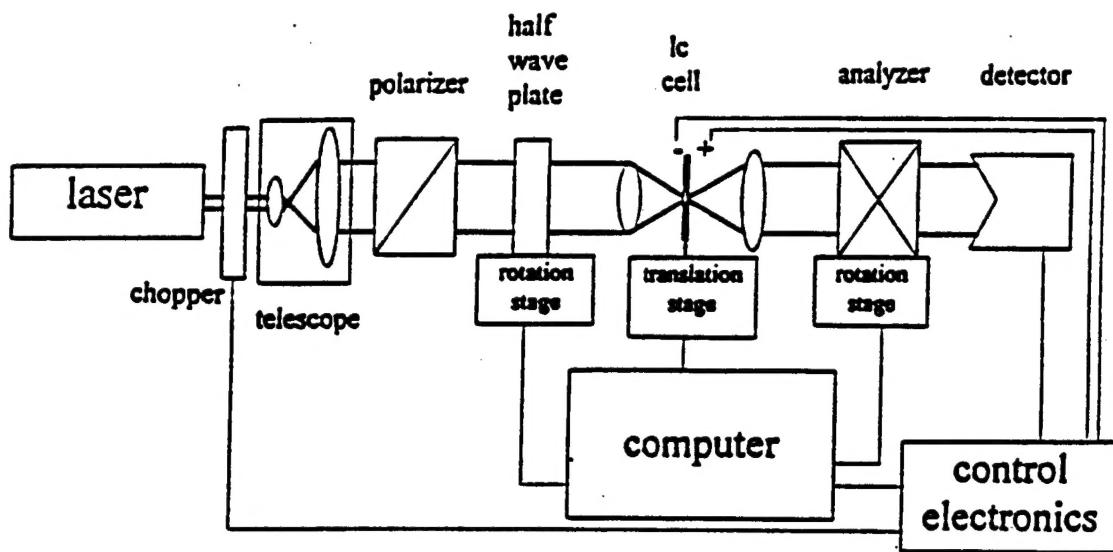


Figure 2. Schematic diagram of the experimental apparatus.

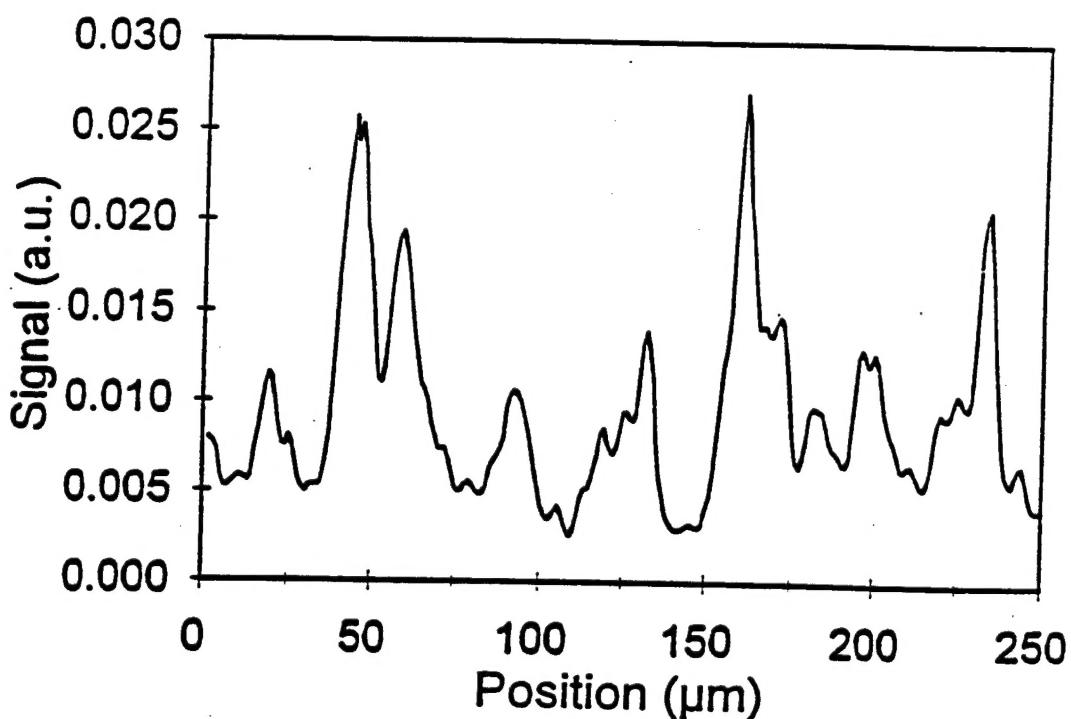


Figure 3. Variation of the signal from a $15\ \mu\text{m}$ KN125 sample between crossed polarizers as a function of the position at $632.8\ \text{nm}$.

The cw laser beam was chopped at 10 Hz with a 3 percent duty cycle. A 10 Hz bipolar square-wave voltage of variable amplitude was applied across the liquid crystal cell and synchronized with the optical beam. This permitted the response of the liquid crystal to electrical signals of either polarity to be interrogated. The variation of the transmitted signal was detected with either a photodiode/lock-in amplifier pair or via a calibrated energy meter.

The dependence of the electro-optical properties of the liquid crystal on electric field, optical polarization and position were studied. Figure 3 shows the cross-polarized transmission of the 15 μm thick sample as the focussed laser spot is scanned along the X direction, perpendicular to the stripe deformation. A quasi-periodic modulation of the transmission associated with the stripe geometry was observed. As shown in the figure, the position of the stripes can be determined with micron resolution, permitting the optical measurements to be made within a given stripe domain. The observed periodicity for the 15 μm thick sample is on the order of 20 μm . These spatially resolved transmission measurements very clearly illustrate deviations from periodicity in the stripe texture that are also observed in published micrographs.^{1,2}

The light transmitted by a birefringent material between crossed polarizers is given⁴ by

$$I_{\perp} = I_0 \sin^2 2\phi \sin^2 \delta/2 \quad (1)$$

where ϕ is the angle between the polarization direction and the layer normal and δ is given by

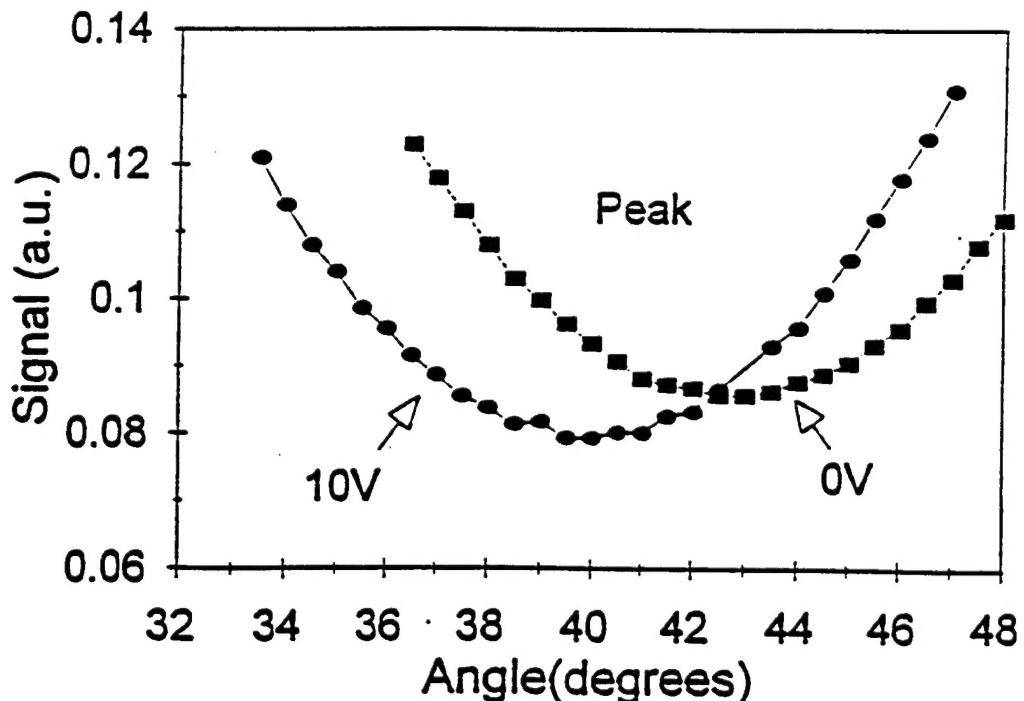


Figure 4. Angular scans at a peak position of the light transmitted through a 15 μm KN125 sample between crossed polarizers at 488.0 nm at different voltages.

$$\delta = \frac{2\pi \Delta n d}{\lambda} \quad (2)$$

with Δn the birefringence, d the sample thickness and λ the wavelength of the light. As one scans from peaks to valleys in the X-scan the transmission varies because the molecular orientation alternates between adjacent stripes in the quasi-bookshelf geometry. Since the term $\sin^2\delta/2$ is independent of X , the position dependence of the transmission enters through variations of the angle ϕ .

The optical beam was next confined to a single stripe domain, i.e., to a peak in the X-scan, and the transmission of the liquid crystal was measured as a function of the angle ϕ between the polarization vector and the molecular director. Fig. 4 presents angular scans for voltages of 0 and 10 V as ϕ is varied by rotating the polarization of the optical beam. These results demonstrate the sensitivity of the transmission to field induced variations in molecular orientation. These may be associated with the electroclinic tilt angle or changes in the orientation of the striped domain. A comparison of the angular position of the transmission minimum for a peak (in the X-scan) with that of an adjacent valley at a fixed voltage provides information on the relative orientation of the layer normals for adjacent stripe domains (see Fig. 5). From the angular shifts in the transmission minimum shown in figures 4 and 5, the true optical tilt angle can be determined for a given stripe domain. Fig. 6 shows the electric field dependence of the tilt angle for KN125 determined directly from spatially resolved electrooptic measurements using 632.8 nm laser radiation. These results agree quite well with independent tilt angle measurements derived using conventional optical and X-ray techniques.¹

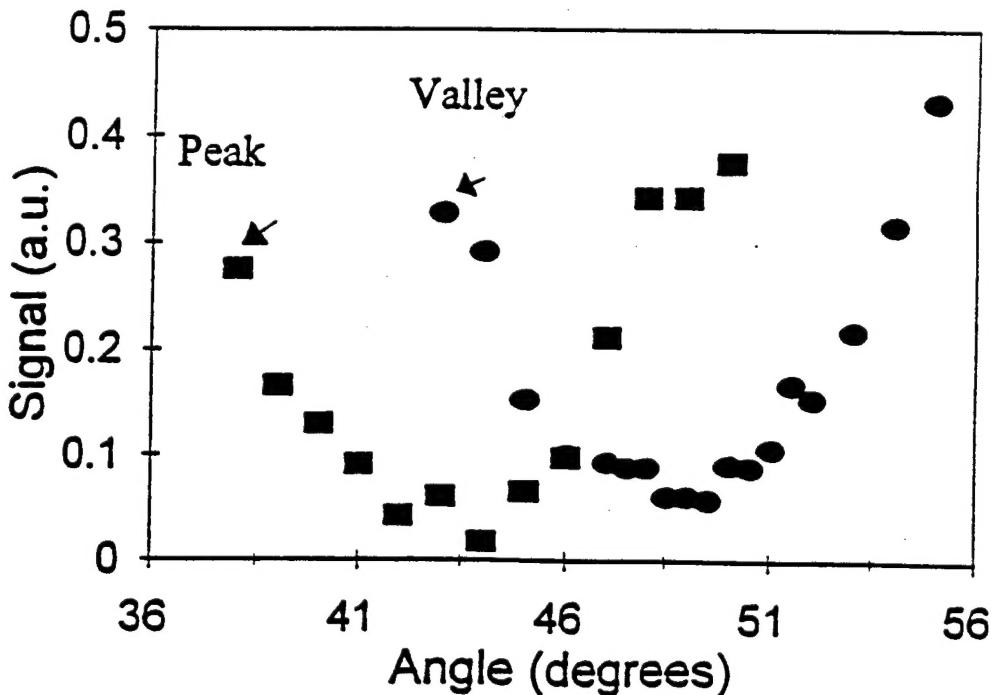


Figure 5. Angular scans of adjacent peak and valley positions at the same voltage. 15 μm KN125 at 488.0 nm.

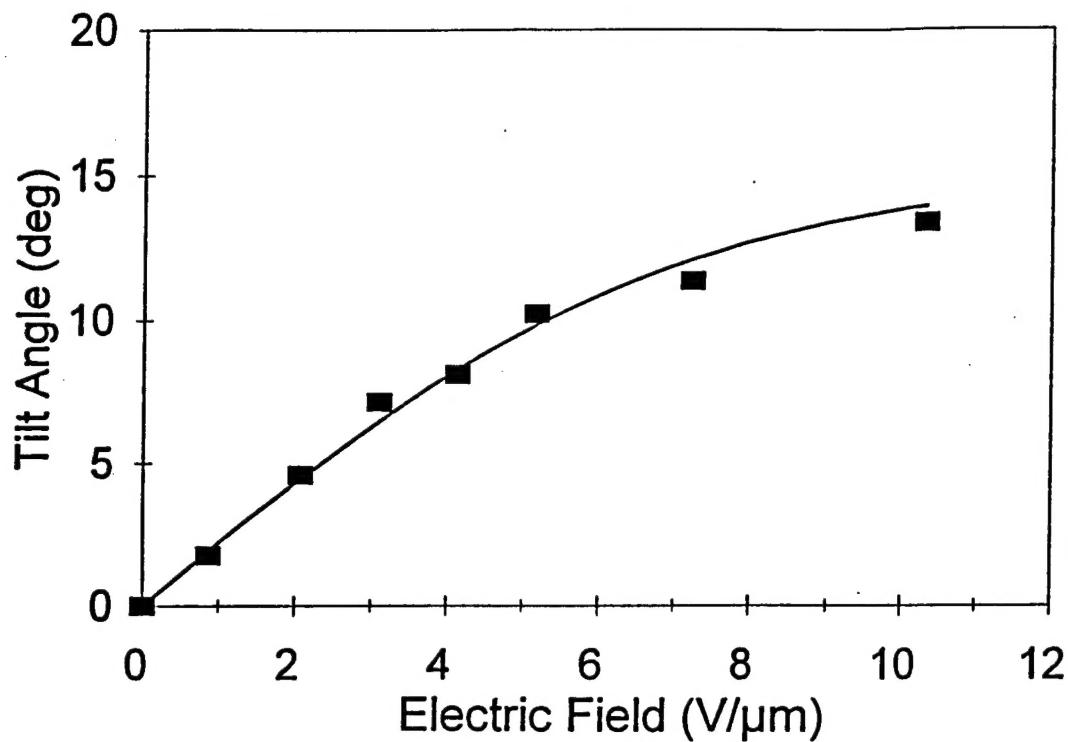


Figure 6. Variation of the tilt angle as a function of field. 15 μm KN125 at 632.8 nm.

In summary, a novel optical technique has been developed to probe the electrooptic properties of liquid crystals with micron-scale spatial resolution. The technique was employed to determine the effect of the stripe deformation on the optical properties of the electroclinic liquid crystal KN125. Measurements of the polarized optical transmission as a function of position within the sample exhibit a quasi-periodic modulation due to stripe deformation of the bookshelf geometry. Electrooptic measurements performed within single stripe domains permitted the true optical tilt angle and the angle of the stripe deformation to be determined. The electric field dependence of the optical tilt angle for the chiral smectic A liquid crystal KN125 was reported.

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